Superfluid density of thin ^{4}He films adsorbed in porous Vycor glass

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We have studied the superfluid density of thin superfluid ^{4}He films absorbed in porous Vycor glass. At low temperatures we have used a third-sound technique to examine the Landau elementary excitations. We observed phonons with a T^2 temperature dependence and rotons with a 5-K energy gap. At higher temperatures we have examined the superfluid density in the critical region. The superfluid density follows a power law similar to that of bulk ^{4}He with an exponent at the bulk value. We conclude that these effects are due to the three-dimensional geometry of the substrate.

INTRODUCTION

The question of long-range order in reduced dimensionality systems has been the subject of considerable interest for many years. For example, Bloch[1] in 1930 showed that one- and two-dimensional magnetic systems cannot have any spontaneous magnetization at a nonzero temperature. In 1935 Peierls[2] showed that crystalline order was also destroyed at any nonzero temperature. More recently, the absence of long-range order of the conventional form has been shown by Mermin[3] to be a general property of two-dimensional systems. However other types of long-range order have been suggested for two-dimensional systems such as topological long-range order suggested by Kosterlitz and Thouless.[4] Therefore there has been considerable and current theoretical interest in the nature of long-range order in reduced dimensionality systems.

Experimentally many systems hold promise for testing the relevance of these theoretical ideas. Monolayers of gas adsorbed on various substrates,[5] two-dimensional electron gases,[6] metallic thin films,[7] layered metallic[8] and magnetic systems,[9] and smectic phases of liquid crystals[10] are examples of such experimental systems. However few systems hold as much promise for high-precision critical-point studies as thin superfluid films. The high purity of sample, availability of Bose or Fermi systems, and the ease with which sample thickness and topology can be altered make the helium film an extremely versatile system.

Unsaturated ^{4}He films have been studied using a variety of experimental techniques. The methods used include mass and thermal[11–14] transport, propagation of third sound,[15,16] and persistent currents.[17] Recently, however, there have been developed several methods which allow the potential of ^{4}He as a high precision reduced dimensionality system to be realized. These include a fourth-sound technique developed by Hall and Reppy,[18] a third-sound technique developed by Rudnick, Mochel, and co-workers (see, for example, Ref. 19), and an Andronikashvili method developed by Reppy and co-workers.[20–23] These techniques have allowed extremely high precision studies to be made on the effects of size, ^{3}He impurity, and dimensionality on the superfluid phase transition. In this paper we would like to describe measurements performed on ^{4}He films adsorbed in porous Vycor glass using two high precision techniques—the Hall-Reppy jiggler and the Andronikashvili torsional pendulum. Vycor glass is useful in this type of experiment because it provides a large continuous surface area ($\sim$100 m^2/g). Also the interior channels of the glass form a highly interconnected three-dimensional network. Vycor glass is preferable because of the extremely uniform distribution of pore sizes in comparison to other systems of this type such as packed powders (for a discussion of this point see Ref. 24). Such uniformity allows very careful studies close to the transition to be made. These studies give insight to the effects of size and reduced dimensionality on the nature of the superfluid transition. The first studies of Vycor glass as a superleak were performed by Atkins, Seki, and Condon[25] in 1956. Since that time many workers have studied ^{4}He adsorbed into Vycor glass including Rudnick and co-workers,[26] Brewer and co-workers,[27,28] and Reppy and co-workers.[17,18,20,29–32] Only recently however has the sharpness of the superfluid transition in this system been realized. Work by Gregory *et al.*,[24] Kiewiet *et al.*,[18] Berthold *et al.*,[20] and Tyler *et al.*[33] have pointed to the value of ^{4}He adsorbed on Vycor as a high precision experimental system to test theoretical ideas such as the effects of size and dimensionality on the nature of the superfluid transition.

In this paper we investigate two separate regions of

the superfluid phase diagram. In Sec. I we discuss low-temperature measurements made using a third-sound hall-Reppy technique. We fit our results to the Landau theory of elementary excitations. In Sec. II we discuss measurements made using an Andronikashvili torsional pendulum and the nature of the superfluid density near the critical temperature. In Sec. III we draw our conclusions.

I. LOW-TEMPERATURE REGION —THIRD-SOUND METHOD

In this section we would like to describe measurements made on superfluid ^{4}He films adsorbed on a porous Vycor glass substrate using a third-sound technique. The technique is due to Hall and Reppy and is described elsewhere.[18,29,34] Using this oscillating cavity technique we have examined third-sound resonances for coverages ranging from 1.5 to 2.7 atomic layers at temperatures ranging from 0.23 to 1.25 K.

He II films are capable of sustaining a unique sound mode called third sound. In this mode the normal fluid component is clamped to the substrate by its viscosity and only the superfluid component of the film moves. That such a sound mode should exist was first theoretically predicted by Atkins[35] in 1959 and experimentally observed by Everitt, Atkins, and Denenstein in 1964.[36] The wave velocity U_3 is given by Atkins[35] as

$$U_3^2 = (\rho_s/\rho) f d \ , \tag{1}$$

where ρ_s/ρ is the superfluid density of the film of thickness d experiencing a van der Waals force f.

The Hall-Reppy "jiggler" technique involves setting up third-sound standing waves in an oscillating cavity. Resonances in the system correspond to fitting an odd number of half-wavelengths into the cavity. In practive only the fundamental mode was used. The equations of motion for this type of cell have been derived elsewhere.[34,37] Only the results are given here. The motion of a Hall-Reppy type of cell is given by

$$X = F_0 e^{i\omega t}\left[K - (M+m)\omega^2 - i\omega D + \frac{m_s}{n^2}\left[1 - \frac{2}{kL}\tan\left(\tfrac{1}{2}kL\right)\right]\left[\frac{\omega}{C_3 k}\right]^2\right]^{-1} , \tag{2}$$

where: X = displacement of cell; $F_0 e^{i\omega t}$ = driving force on cell; K = spring constant of cell; D = damping of cell; M = total mass of cell; m = mass of fluid in cell; m_s = superfluid mass; n = index of refraction of system; C_3 = third-sound velocity; $k = \omega C_3^{-1}(1 + iQ_3^{-1})^{1/2}$; and $Q_3 = \omega\rho_s\eta^{-1}$.

A fit to the data using (2) can be made yielding the quantity (m_s/n^2) from which the superfluid mass m_s can be extracted.

The cavity (shown in Fig. 1) consists of a piece of Vycor glass (Corning porous glass 7930), potted in Stycast 1266 epoxy and supported by a drill rod suspension. The cavity is driven by a capacitor on one end and its motion is detected by a microphone on the other end. The frequency of oscillation of the cavity is varied and third-sound resonances are observed as sharp peaks in the velocity of oscillation.

In operation, first the frequency spectrum of the empty cell is examined. Then ^{4}He is condensed into the cell and the frequency spectrum is reexamined. In this fashion the principle third-sound standing wave mode is identified. The temperature is then varied and the frequency of the mode is measured as a function of temperature. Shown in Fig. 2 is a plot of this third-sound principle mode-resonance frequency as a function of temperature for two different thickness films. Unfortunately as one approaches to within ~50 mK of the transition, the resonances become strongly damped and one cannot follow them into the "critical region."

The piece of Vycor in the third-sound cell is 1.5 cm long. Using the third-sound resonant frequency and an appropriate index of refraction, one can calculate a third-sound velocity for our films. Using an Andronikashvili oscillator[20] we have obtained $n = 1.15$ for a similar piece of Vycor. For $n = 1.15$ we find that the 31.5 μmole/m^2 ^{4}He film has a third-sound velocity of 41.4 m/sec at $T = 0$.

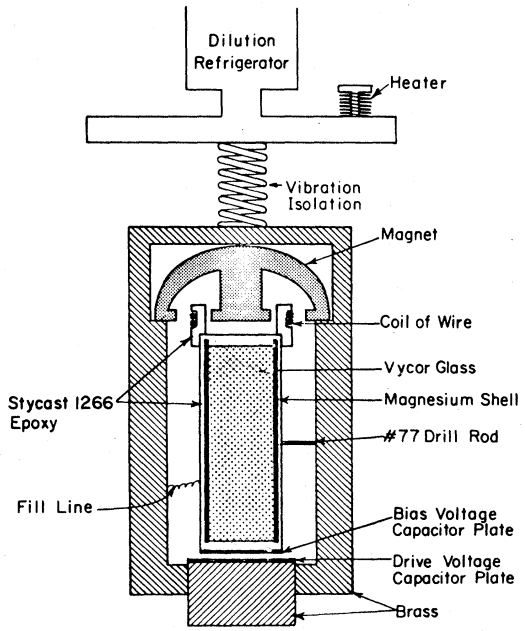

FIG. 1. The third-sound experimental cell is shown.

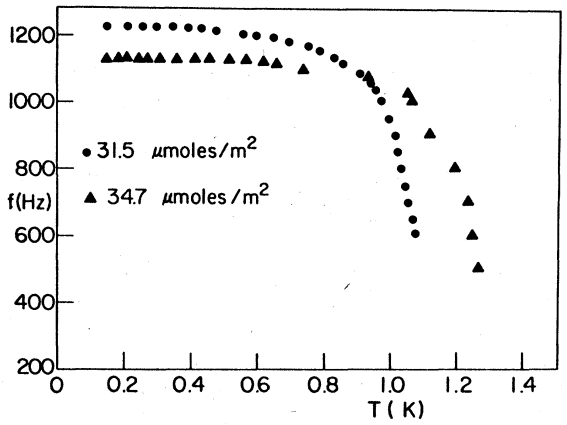

FIG. 2. Third-sound resonance frequencies for two films are shown as a function of temperature.

In order to avoid the necessity of using a model-dependent index of refraction to analyze our data we use the following relation to calculate $\rho_s(T)/\rho$ from the third-sound resonant frequencies:

$$\rho_s(T)/\rho = [U_3(T)/U_3(T=0)]^2 = [f_3(T)/f_3(T=0)]^2 \ . \quad (3)$$

Using Eq. (3) we have obtained ρ_s/ρ data as a function of temperature and film thickness.

A representative sample of this $\rho_s(T)/\rho$ data is shown in Fig. 3. The thinnest films studied using this third-sound technique had a transition temperature of 0.65 K and the thickest films (full Vycor pores) has a transition temperature of 1.955 K. For comparison the bulk superfluid density is also shown in Fig. 3. As regards Fig. 3, several points should be noted. As the films become thinner, the onset temperatures decrease. Also ρ_s/ρ in the films falls off from the value at $T=0$ more quickly than the bulk value of ρ_s/ρ.

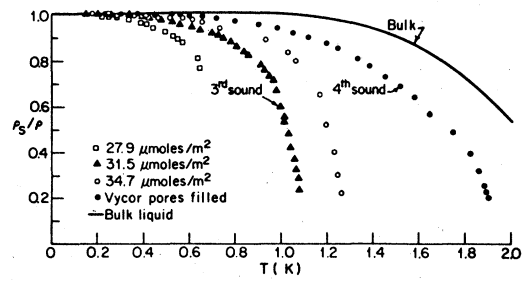

FIG. 3. Shown are the superfluid densities for ^{4}He films adsorbed in Vycor glass as determined using the third-sound technique.

In Fig. 4 we examine the onset temperatures as a function of total film thickness. Note the linear dependence and the nonzero intercept. We have also plotted the superfluid masses using a fit of Eq. (2) to the third-sound resonances. If we average the intercepts we obtain 21.8 μmole/m^2 or 1.35 monolayers as the thickness of the nonsuperfluid layer. This value represents a minimum film thickness for superflow or the thickness of helium required to support superflow. Such "solid" layers have been observed in numerous experiments on both Vycor glass[17,20,30] and other substrates.[38,39] Our numerical value is in rough qualitative agreement. All of our measurements including both third-sound and Andronikashvili[20] methods have indicated that the superfluid in excess of this solid or nonsuperfluid layer acts like a dilute surface gas. Using an Andronikashvili cell we have examined superfluid films with a thickness of 0.01 active atomic layers. We see no evidence of a percolation-type transition[40] in helium films, even when the amount of active superfluid is as little as 0.01 atomic layers.

In order to understand the low-temperature regime of our data we have adopted the picture of Landau's elementary excitations.[41] In this picture the thermally excited surface phonons and rotons form a normal fraction which is clamped to the substrate by its viscosity. This idea has been used to explain the behavior of bulk ^{4}He below 1 K. Its application to systems of reduced dimensionality was discussed theoretically by Kuper,[42] Padmore,[43,44] Padmore and Reppy,[45] and Haug,[46] and Götze and Lücke.[47] Experimental analysis of superfluid film data within this framework has been performed by Brewer,[27], Bishop, Parpia, and Reppy,[29] Smith, Bishop, Berthold, and

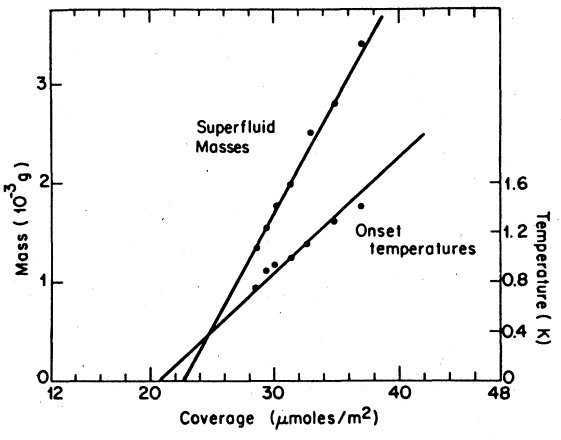

FIG. 4. Shown are the onset temperatures and superfluid masses as a function of film thickness for ^{4}He films adsorbed in Vycor glass as measured using the third-sound technique.

Reppy[31]; Washburn, Rutledge, and Mochel,[48] Chester and Eytel,[49] Kiewiet, Hall, and Reppy,[18] and a very complete discussion has been given by Rutledge, McMillan, and Mochel.[19] In this analysis we assume that the normal fluid is comprised of a gas of roton and phonon excitations whose character is modified by the reduced dimensionality of the system.

In the temperature range below 0.5 K one expects the normal fluid component to be due solely to phonon-type excitations; the rotons will be "frozen out." The low-temperature behavior of our ρ_s/ρ data conforms to this picture. For the temperature range below 0.5 K we find that the superfluid density follows a T^2 temperature dependence. This is shown in Fig. 5 where ρ_s/ρ is plotted versus T^2 and very good straight lines are obtained.

Padmore,[44] and Cole and Saam[50] have provided the theoretical framework within which to understand this result. In Padmore's calculation the phonon contribution to the normal fluid density in systems of various dimensionality is derived. Padmore concludes that the temperature dependence of the phonon term in the normal fluid density should go as T^{D+1} where D is the dimensionality of the system. In his model a system of flat plates is two dimensional, cylindrical tubes are one dimensional, and packed powders are described as being "zero dimensional." In his picture the cylindrical Vycor pores would be expected to have a T^2 normal fluid density phonon term. Studies of ^{4}He films in systems of planar geometry provide a check of this interpretation. Indeed, third-sound studies by Rutledge *et al.*[19] on a flat plate geometry do confirm Padmore's predictions and observe a T^3 phonon term.

In the calculation of Cole and Saam,[50] the hydrodynamics of superfluid helium confined to cylindrical channels was studied. They also predicted the existence of a phonon mode with a T^2 temperature dependence. In addition they predicted that at certain critical filling factors, partially filled cylindrical pores, would be unstable and fill spontaneously. Such instabilities have been observed by Chen *et al.*[51]

In addition to the phonon contribution to ρ_s/ρ we also find a roton contribution at temperatures greater than 0.5 K. To analyze our data for rotons we subtract off the low temperature T^2 phonon contribution and fit the remainder to a roton contribution term of the following form:

$$\rho_n(T) \sim T^{-1/2}\exp(-\Delta/kT) \tag{4}$$

from this analysis we are able to obtain a roton energy gap Δ.

In Fig. 6 is shown an analysis of this type. The quantity plotted is the reduced normal fluid density minus the phonon contribution. This quantity plotted versus $1/T$ should yield a straight line if a temperature-independent roton gap contribution can be used to explain deviations from a phonon contribution to the normal fluid density. Obviously a very good fit can be obtained.

The roton energy gaps obtained in this analysis are shown in Fig. 7. Also shown are values obtained by other workers for helium adsorbed on different substrates. The data points by Rutledge *et al.*[19] are obtained on a two-dimensional substrate of crystallized argon. The points due to Brewer *et al.*[27] and Kiewiet *et al.*[18] are also obtained using porous Vycor glass as a substrate. The point due to Chester *et al.*[49] was a fit by them to data obtained by Scholtz *et al.*[38] on a CaF_2 single-crystal substrate. The values of the energy gaps are in the range of 3–6 K. These values are substantially lower than the bulk value of 8.65 K and appear to be independent of film thickness over this range of films.

Recently Thomlinson *et al.*[52] have used inelastic neutron scattering from ^{4}He adsorbed on Graphon to directly observe the two-dimensional roton gap.

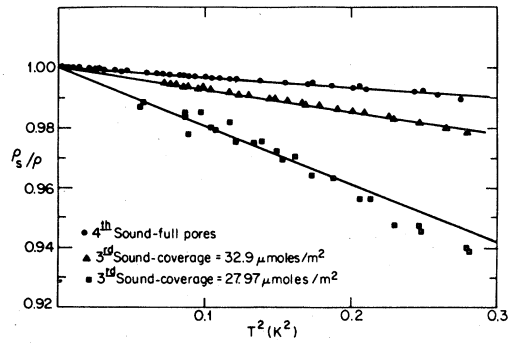

FIG. 5. Shown is the low-temperature T^2 dependence of the superfluid density of ^{4}He adsorbed in porous Vycor glass.

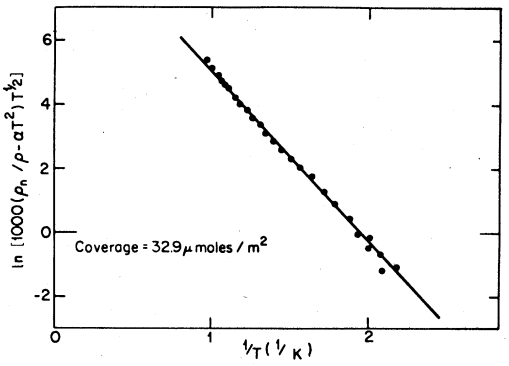

FIG. 6. The reduced superfluid density is shown as a function of inverse temperature. The straight line corresponds to a temperature-independent roton contribution with a gap of 4.9 K.

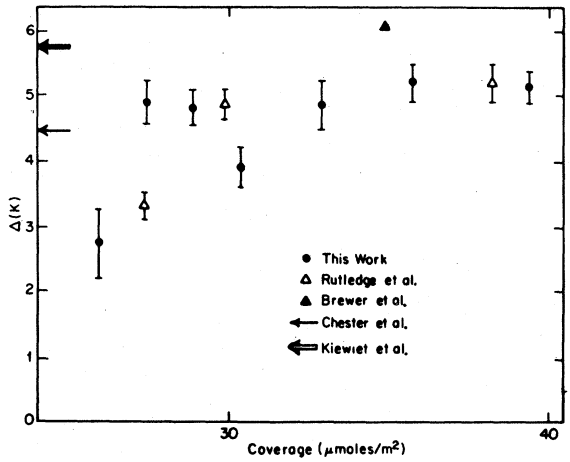

FIG. 7. The roton gap is shown as a function of film thickness from this work and as determined by other workers.

Their results are consistent with these earlier thermodynamic observations.

Several workers have theoretically addressed the question of reduced roton gaps in reduced dimensionality systems. The most straightforward analysis was suggested by Brewer, Symonds, and Thomson.[27] Their idea is shown schematically in Fig. 8. It has been observed in the bulk that the roton gap is a function of density. The bulk roton gap data shown in Fig. 8 are from the bulk entropy measurements by van den Meijdenberg, Taconis, and de Bruyn Ouboter.[53] By extrapolating to the value of 5.85 K for the roton gap obtained by Kiewiet *et al.*[18] for full pore Vycor glass, one obtains a density of 0.188 g/cm^3. This compares very well with the value of the second layer density of 0.183 g/cm^3 obtained by

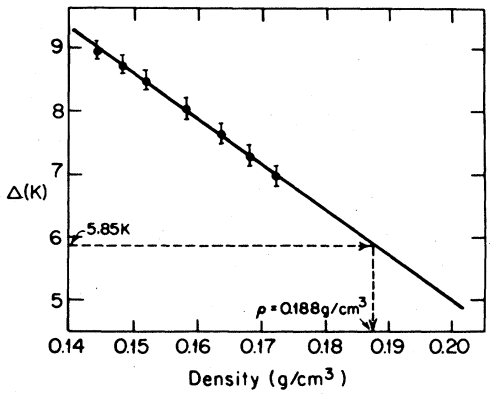

FIG. 8. The dependence of the bulk roton gap on the density as measured by van den Meijdenberg, Taconis, and de Bruyn Ouboter. The dotted line corresponds to the gap as determined for ^{4}He in Vycor glass.

Brewer[27] for helium in porous Vycor glass. This simple analysis would indicate that the reduced value roton gap is due to the second dense layer of the helium adsorbed on the porous Vycor glass. This model would also explain why the gaps appear to be independent of coverage since the second-layer density is roughly independent of total coverage.

Padmore[43] has given a similar answer as a result of a more sophisticated calculation. Padmore performed a Monte Carlo calculation of the Feynman-Cohen excitation spectrum for helium confined to two dimensions. He finds that the roton gaps he obtains are substantially lower than the bulk values and are in rough agreement with experiment at the appropriate densities.

In an early work Kuper[42] in 1958 calculated the effect of a wall on the reduction of the roton energy. He gives a hydrodynamic argument which indicates that the roton energy gap for a roton near a wall should be reduced by about a factor of 2. As pointed out by Kuper the validity of using hydrodynamics at this sort of scale is open to question. However he did obtain a result in agreement with both later theory and experiment.

Most recently there has been work on this problem by Götze and Lüke.[47] They estimate the spectrum of rotons in two-dimensional liquid helium II within the Bijl-Feynman theory. They indicate that due to stronger short-range order and more effective back flow, the roton gap is reduced by a factor of 2 from the bulk value.

Therefore there seems to be considerable theoretical agreement on this problem. There appear to exist two-dimensional rotons which dominate the behavior of ρ_s/ρ in this region.

The following picture emerges from this analysis. The phonon excitations are long-wavelength objects. The system geometry is important in determining their character. Therefore one sees different phonon behavior in planar or cylindrical geometries. However the rotons are small objects. Their character is modified by the presence of the wall but not by the system geometry. Hence the agreement among measurements for roton gaps in systems of different geometry. The values of the roton gaps are reduced by wall effects. However they are independent of film thickness (or in Vycor, fractional filling) as observed by experiment. Presumably for rotons it is the solid-liquid interface in the film which is important.

In the final figure (Fig. 9) of this section we show our data for the entire temperature range investigated. The theoretical curves are calculated using the combination of a T^2 phonon term and a roton term. At higher temperatures quasiparticle interactions which have been ignored in this simple Landau theory presumably become important and worsen the agreement with the data. However below approxi-

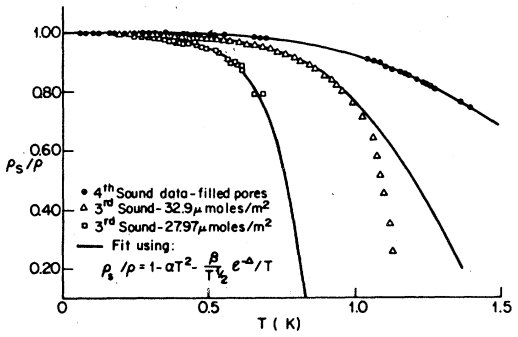

FIG. 9. Shown in the superfluid density for ^{4}He adsorbed in Vycor glass as measured using the third-sound technique. The theory curves are discussed in the text.

mately 1 K where one would expect such an analysis to be valid, agreement with the data is very good.

Therefore we conclude that the Landau picture of elementary excitations is adequate to describe the low-temperature behavior of the superfluid density of ^{4}He films, provided one properly modifies their character to take into account the dimensionality and surfaces in the system. As mentioned earlier the simple picture of noninteracting elementary excitations becomes invalid at high temperatures when the excitation densities become large. No simple theory seems to be able to explain the data in this region. However, as one approaches the critical region (within $\sim$100 mK of T_c) then one can use ideas that are applicable near the critical point of a system. Unfortunately the third-sound experiment technique fails us as we approach the critical region. The third-sound signals become heavily damped and are difficult to follow close to T_c. In order to examine the critical region we need to develop another method of measuring the superfluid density which will allow us to follow the density through the critical region. Such a technique is described in the next section.

II. CRITICAL-POINT REGION —ANDRONIKASHVILI METHOD

As mentioned above third sound is an inappropriate probe for use in the critical region of a ^{4}He film because as one approaches T_c the third-sound resonances become heavily damped. To overcome this problem we have developed a method for measuring the superfluid density of thin ^{4}He films using a modification of a technique due initially to Andronikashvili.[54]

The Andronikashvili technique makes use of the two-fluid nature of superfluids. In his original experiment a stack of disks at the end of a torsional fiber is placed in a bath of superfluid helium. The plates are closely spaced such that the normal fluid is clamped to them and dragged by them as they oscillate. The superfluid is of course not affected by the plates. By measuring the resonant frequency of the system one can determine the amount of normal mass that is trapped by the plates and determine the superfluid density of the liquid.

Our modification involves using an extremely-high-Q torsional oscilator.[20] By using an oscillator with a $Q \gtrsim 5 \times 10^5$, we are capable of resolving the moment of inertia of the system to better than 1 part in 10^8. Thus, even though we have an extremely thin ^{4}He film (a few atomic layers) we have enough resolution to allow us to resolve the superfluid mass of these films to a part in 10^5. This technique of course allows us to follow the superfluid density through the transition region. With proper temperature control we are able to make an extremely precise determination of the superfluid density for ^{4}He films in their critical regions. Prior to the present work, only the quartz microbalance technique developed by Chester and Yang[55] provided a measure of the superfluid mass through the transition region. Other methods using heat transfer, mobility, third sound, or measurement of persistent current angular momentum suffered from a loss of singular or difficulty in interpretation as the critical temperature was approached.[28] Our method is similar to that used by Guernsey *et al.*[56] at Columbia and Main *et al.*[57] at Manchester and has been described elsewhere.[20–22,58]

In our cell (shown in Fig. 10) a cylinder of porous Vycor glass is incorporated into the bob of a torsional pendulum with a resonant frequency of approximately 1570 Hz. The helium for the experiment is admitted through the hollow torsion rod. The oscillator is

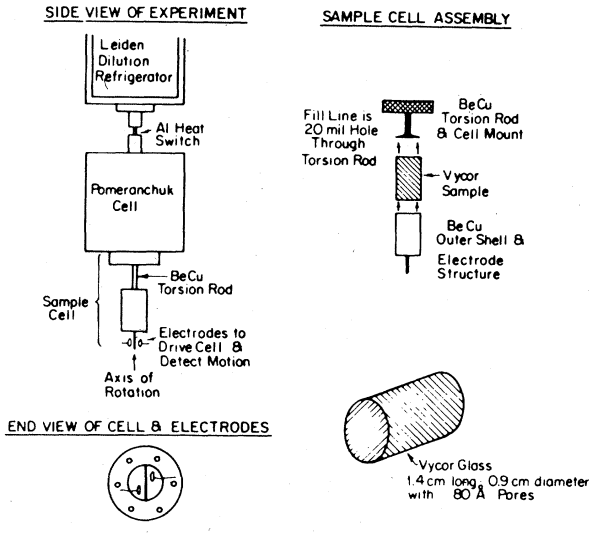

FIG. 10. Shown is the construction of the Vycor Andronikashvili cell.

driven and its motion detected electrostatically. The oscillator is incorporated into a feedback loop consisting of an amplifier, active filter, phase shifter, and zero-crossing detector.[32] In such a circuit the oscillator is the frequency-determining element. Hence by simply counting the resonant frequency of the loop we can determine the moment of inertia to 1 part in 10^8.

The analysis of the data assumes that all of the normal fluid is locked to the walls of the system. Above the transition, the fluid contributes its entire moment of inertia to the system. Therefore the fractional filling of the Vycor pores can be monitored by measuring the frequency shift, above the transition temperature, as helium is admitted. The total fractional shift in frequency between empty and full pores is 3.06×10^{-3}.

In operation, helium is admitted into the cell and the cell allowed to anneal at a high temperature. This gives the helium an opportunity to distribute itself uniformly. The cell is then cooled and the period $P(T)$ is measured as a function of temperature. At each point the temperature is regulated to ± 10 μK and the period is allowed to stabilize to several parts in 10^9 before a reading is taken. Equilibrium time between points is approximately 30 min. When the oscillator is cooled below the transition temperature, T_c, superfluid is formed. The previously constant period, $P(T)$, decreases with temperature. The quantity of interest is the shift in period $\Delta P(T)$:

$$\Delta P(T) = P(T_c) - P(T) \ . \tag{5}$$

The superfluid mass is proportional to $P(T_c)^2 - P(T)^2$. However because the fractional shift is small, the superfluid mass is closely proportional to $\Delta P(T)$. We have measured this quantity $\Delta P(T)$ for films with transitions ranging from 1.5 K to 25 mK. The results are shown in Fig. 11.

In addition to measuring the period of oscillation of the system we can also measure the amplitude as a function of temperature for a fixed drive. Because the oscillator has an extremely high Q, this measurement of the amplitude becomes an extremely sensitive measure of the dissipation of the system. This method was used for example to measure the dissipation of a two-dimensional Kosterlitz-Thouless superfluid[23] on a Mylar film. However in the current experiment we have seen no dissipation accompanying the superfluid transition. This we take as being due to the three-dimensional nature of our Vycor substrate. This experiment in conjunction with the two-dimensional Mylar experiments[23] we feel provide convincing evidence for the importance of the role that substrate dimensionality plays in the nature of the superfluid transition of ^{4}He films.

In contrast to the original Andronikashvili experiment performed with perfect parallel plates, the superfluid part of the film adsorbed on the Vycor glass

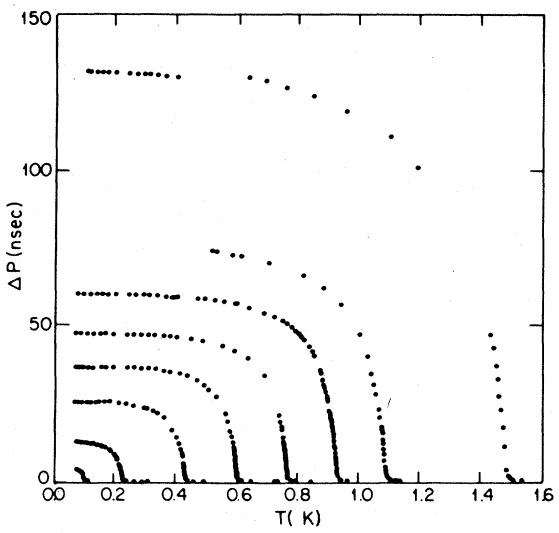

FIG. 11. The difference between the period at the transition temperature, T_c, and the period at a temperature T, $\Delta P(T)$, is plotted as a function of T for a number of different thickness films.

must execute a complex potential flow as the pendulum oscillates. As a result, in Vycor a fraction, χ, of the superfluid moment of inertia, I_s, remains effectively locked to the pendulum,[59] and the observed period shift is proportional to the unlocked fraction $(1-\chi)I_s$. To calculate the superfluid density ρ_s/ρ we use a normalization at $T=0$ to eliminate this geometric flow factor

$$\rho_s/\rho = \Delta P(T)/\Delta P(0) \ , \tag{6}$$

where $\Delta P(0)$ is obtained by extrapolation to $T=0$.

In Fig. 12 we show the measured values of $\Delta P(0)$ and T_c as a function of fractional filling σ of the Vycor. Both quantities show a linear dependence on fractional filling with a nonzero intercept σ_0. σ_0, the intercept, is the same for both quantities to within 1%. Thus the Andronikashvili measurement shows the same qualitative behavior as does the third-sound technique. This linear dependence of $\Delta P(0)$ and T_c on coverage contradicts the existence of a percolation-type transition in any of the films we have studied.

In addition we have examined the phase diagram in the pressure-temperature plane for ^{4}He adsorbed in Vycor glass. These results are shown in Fig. 13. For this experiment the Vycor pores are filled with ^{4}He at the appropriate pressure. The period of the Vycor Andronikashvili cell was used to indicate the onset of superfluidity as a function of temperature. In this fashion the "lambda" line for helium confined to porous Vycor glass was mapped out as a function of pressure and temperature. As shown in the figure the slope of this line is equal to that of the bulk

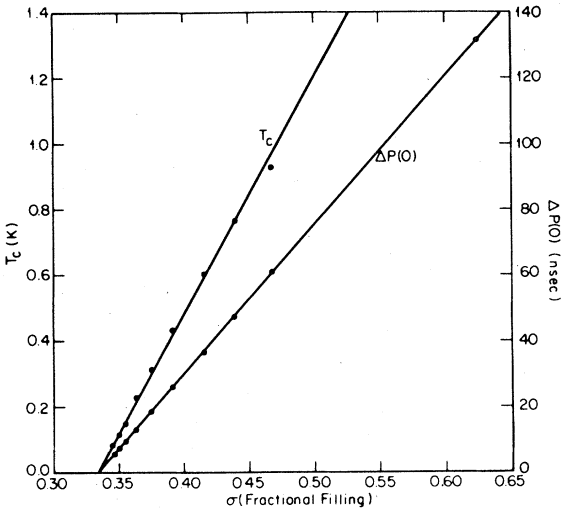

FIG. 12. The transition temperature and $T=0$ period shift as measured by the Andronikashvili cell are shown as a function of fractional filling.

lambda line. If the simple interpretation of the pores contributing a net pressure to the system (~14 bars) were applicable then one would expect to see superfluid flow blocked by solid formation at a pressure well below the bulk melting pressure. This is not observed.[60] Therefore a much more complicated explanation is required than we have at the moment.

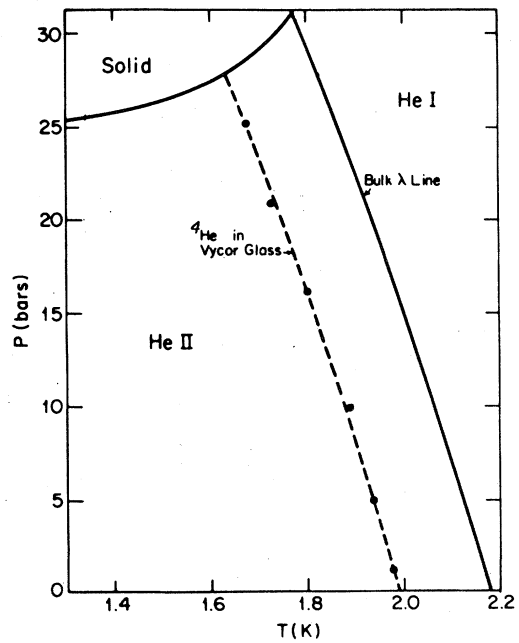

FIG. 13. The lambda line as a function of pressure is shown for ^{4}He-filled Vycor pores.

During all of our runs with the Andronikashvili cell, third-sound resonances were observed. These occurred when various third-sound modes had frequencies which matched the resonant frequency of the cell. The gaps in the data in Fig. 11 correspond to the locations of these third-sound modes. These modes pulled the resonant frequency of the oscillator at low temperatures making the data useless for the evaluation of elementary excitations as was done in the first section. However certain of the modes could be identified and followed as a function of coverage. These modes allowed us to make an interesting correspondence between the third-sound experiments and the Andronikashvili experiments.

In the low coverage regime we expect σ to be proportional to the coverage. The temperature and coverage variation of the third-sound velocity $U_3(T,\sigma)$ is then given by[35]

$$U_3^2(T,\sigma) \sim \left(\frac{3\alpha}{\sigma^4}\right)\left(\frac{\rho_s}{\rho}\right)(\sigma-\sigma_0) \ , \tag{7}$$

where α is the van der Waals coefficient.

Because the superfluid mass ρ_s/ρ in (7) is proportional to $\Delta P(T,\sigma)$ we may rewrite (7) as

$$U_3^2(0,\sigma) = U_3^2(T,\sigma)\left(\frac{\Delta P(0,\sigma)}{\Delta P(T,\sigma)}\right) \ . \tag{8}$$

From (7) we can also see that the zero-temperature value $U_3^2(0,\sigma)$ is proportional to $[(\sigma-\sigma_0)/\sigma^4]$. This function has a maximum at $\sigma=\frac{4}{3}\sigma_0$. Therefore we may write

$$\frac{U_3^2(0,\sigma)}{U_{3\max}^2} = \frac{U_3^2(0,\sigma)}{U_3^2(0,\frac{4}{3}\sigma_0)}$$

$$= \left(\frac{\sigma-\sigma_0}{\sigma^4}\right) \Big/ \left(\frac{\frac{4}{3}\sigma_0-\sigma_0}{(\frac{4}{3}\sigma_0)^4}\right)$$

$$= \left(\frac{256}{27}\right)\left(\frac{\sigma-\sigma_0}{\sigma^4}\right)(\sigma_0^3) \ . \tag{9}$$

In Fig. 14 we have plotted $U_3^2(0,\sigma)/U_{3\max}^2$ from three different sources. The open circles are the third-sound resonances as seen in the Andronikashvili experiment and calculated using Eq. (8). Each point represents an average value obtained from the observation of several cavity modes at each coverage. The solid circles represent the same quantities as measured directly using the third-sound technique described in Sec. I. The solid line is Eq. (9) using $\sigma_0=0.337$, the value obtained from the $\Delta P(0)$ and T_c versus coverage data. Note that although the third-sound data do not extend to as low coverages, there is good agreement among all three. The high coverage region shows $U_3^2(0,\sigma)/U_{3\max}^2$ rising again.

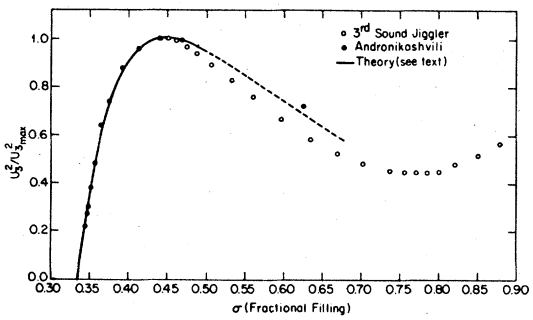

FIG. 14. The ratio of third-sound velocity to maximum third-sound velocity at $T=0$ is shown as a function of fractional filling.

This is due to the filling of the Vycor pores and the appearance of a mixed third–fourth-sound mode in the partially filled Vycor pores.

We feel that these two experiments give support to the picture of the superfluid as a dilute surface gas. The existence of sound modes which are standing waves over the entire length of the cavity indicates that superfluidity extends throughout the entire cell and not in just isolated regions. These experiments have been performed at coverages corresponding to 0.01 active monolayers and we see no breakdown of the physically appealing picture of the superfluid as a dilute surface gas adsorbed on a substrate of localized ^{4}He atoms.

The precision of the Andronikashvili method makes itself felt in the critical-region studies for our ^{4}He films. Previous third-sound studies of helium adsorbed on Vycor glass have indicated that the superfluid transition is extremely sharp.[18,20,24,33] Kiewiet *et al.*[18] found that the superfluid density was found to vary near the transition temperature as $(T_c-T)^{\zeta}$ with $\zeta=0.65\pm0.03$. The sensitivity of the Andronikashvili cell has allowed us to examine with high precision the transition region of ^{4}He films with transitions as low as 25 mK. All of our films display a superfluid density, which apart from a small rounding of width $\sim 5\times10^{-3}T_c$, may be characterized by a power law of the form (see Fig. 15):

$$\Delta P(T)\sim(1-T/T_c)^{\zeta}\ . \tag{10}$$

To choose the best values of ζ and T_c the following algorithm was used. The data were linearized according to a power law, i.e., plots of $[\Delta P(T)]^{1/\zeta}$ vs T were made, for various values of ζ. The resultant lines were fit using a linear least-squares-fit routine and the standard deviation Σ was calculated. A plot of the standard deviation Σ vs ζ was made for each coverage (see Fig. 16). ζ was chosen for each coverage as the value which gave the minimum standard deviation Σ. The error bars for each value of ζ were chosen as the 2Σ points. A plot of the values of the

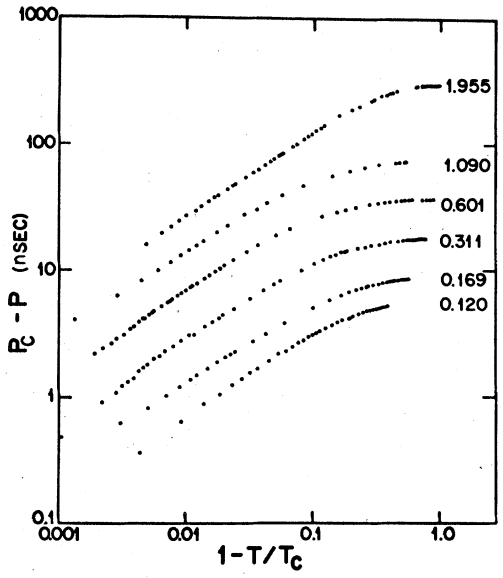

FIG. 15. The change in period $P_c-P(T)=\Delta P(T)$ is plotted against the reduced temperature $1-T/T_c$, for films with transition temperatures ranging from 1.090 to 0.120 K. Full-pore fourth-sound data with a transition temperature of 1.955 K are shown for comparison (Ref. 18).

exponents is shown in Fig. 17. The most important feature is that the nature of the transition is independent of the fractional filling of the pores. Films with transition temperatures ranging from 25 mK to 2 K have the same value of superfluid density exponent. The values of exponents obtained in this experiment range from 0.59 to 0.67 but are uncorrelated with the fractional filling. The average value obtained in this experiment is $\zeta=0.635\pm0.050$. This is to be compared with the value 0.65 ± 0.03 obtained using a

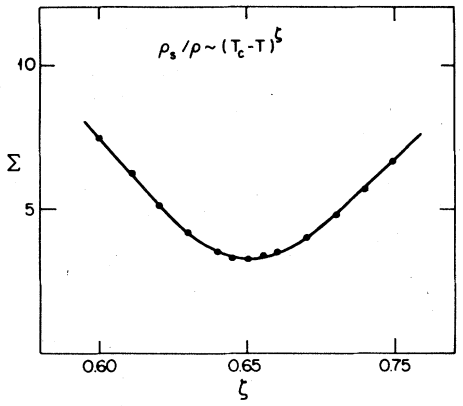

FIG. 16. The standard deviation is shown as a function of the power-law exponent for the superfluid density of a ^{4}He film adsorbed in Vycor glass. The best exponent chosen for each film is that which gives the minimum value of Σ.

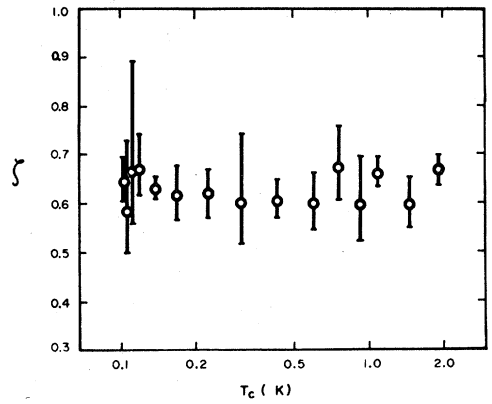

FIG. 17. The superfluid density exponent is shown as a function of transition temperature for ^{4}He films adsorbed in Vycor glass.

fourth-sound technique[18] and the value 0.674 obtained in bulk ^{4}He.[61] This is an extremely surprising result and appears to be due to the three-dimensional character of the substrate. For example, with other studies of ^{4}He films on flat substrates researchers have observed an abruptly changing superfluid mass at onset.[23,39,62,63] The superfluid density in Vycor glass appears to act as though it were a dilute three-dimensional-surface gas. This interpretation of our result is strengthened by recent studies by Bishop and Reppy[23,32] on the superfluid density of thin ^{4}He films adsorbed onto a two-dimensional Mylar substrate. Those results show the behavior of a Kosterlitz-Thouless *two-dimensional* superfluid. These results are radically different from the present results for a three dimensionally interconnected substrate. We feel that these experiments, taken in conjunction, point out the crucial role substrate geometry plays in determining the nature of the superfluid transition in ^{4}He film systems.

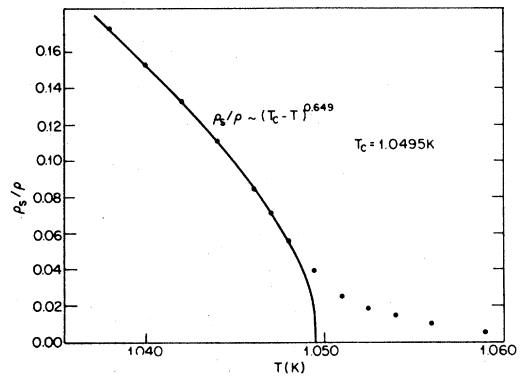

FIG. 18. The region of obvious rounding is shown at the superfluid transition for a ^{4}He film adsorbed in Vycor glass. The solid line is the best power-law fit excluding the rounded region as described in the text.

In this final section we would like to discuss the rounded region at the transition mentioned earlier. In our fits to the power-law behavior this region of obvious rounding was omitted. This region is shown in Fig. 18. The extent of this rounded region depends only weakly on coverage. The value of reduced temperature at which the data depart from a power-law behavior is 0.0015 for a transition at 1.0 K and increases to 0.005 for a transition at 0.1 K and stays roughly constant with a width of ~0.005 K at 25 mK.

In an effort to understand the source of this rounding we have fitted the data using a model of substrate pore size inhomogeneity. It is a well known feature that the transition temperature depends on the pore size in systems of reduced dimensionality. If there existed a distribution of pore sizes then this would produce a rounded transition region similar to what we have seen. Even though we can qualitatively fit our data to such a model, it remains speculative at best to consider whether such a model accounts for our observations. Further experiments on a different piece of Vycor glass with a different pore size distribution are required to sort out the source of this rounded region.

III. CONCLUSIONS

In conclusion we have performed experiments on the superfluid density of ^{4}He films adsorbed in porous Vycor glass using two different techniques. At low temperatures the results can be explained by the use of Landau elementary excitations with modifications from the bulk form due to substrate geometry. We observe phonons with a T^2 temperature dependence and rotons with a 5 K energy gap. At high temperatures in the critical region we observe a superfluid density that follows a power law in reduced temperature similar to that of bulk ^{4}He with an exponent in numerical agreement with the bulk value. We interpret our results as indicating that the superfluid film is a dilute surface gas adsorbed on a substrate of localized atoms. The critical behavior depends crucially on the dimensionality of the substrate geometry. The three-dimensional character of the substrate produces a dilute three-dimensional surface gas, in contrast to experiments performed in a two-dimensional geometry.

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[1]F. Bloch, Z. Phys. 61, 206 (1930).
[2]R. E. Peierls, Ann. Inst. Henri Poincare 5, 177 (1935).
[3]N. D. Mermin, Phys. Rev. 176, 250 (1968).
[4]J. M. Kosterlitz and D. J. Thouless, J. Phys. C 5, L124 (1972).
[5]J. G. Dash, *Films on Solid Surfaces* (Academic, New York, 1975).
[6]M. W. Cole, Rev. Mod. Phys. 46, 451 (1974).
[7]W. J. Skocpol and M. Tinkham, Rep. Prog. Phys. 38, 1049 (1975).
[8]J. A. Wilson, F. J. Di Salvo, and S. Mahajan, Adv. Phys. 24, 117 (1975).
[9]L. J. de Jongh and A. R. Miedema, Adv. Phys. 23, 1 (1974).
[10]P. G. DeGennes, *The Physics of Liquid Crystals* (Oxford University Press, New York, 1974).
[11]E. Long and L. Meyer, Phys. Rev. 79, 1031 (1950).
[12]R. Bowers, D. F. Brewer, and K. Mendelssohn, Philos. Mag. 42, 1445 (1951).
[13]E. Long and L. Meyer, Phys. Rev. 98, 1616 (1955).
[14]D. F. Brewer and K. Mendelssohn, Proc. R. Soc. London Ser. A 260, 1 (1961).
[15]K. R. Atkins, Phys. Rev. 113, 962 (1959).
[16]I. Rudnick, E. Guyon, K. A. Shapiro, and S. A. Scott, Phys. Rev. Lett. 19, 488 (1967).
[17]M. H. W. Chan, A. W. Yanof, and J. D. Reppy, Phys. Rev. Lett. 32, 1347 (1974).
[18]C. W. Kiewiet, H. E. Hall, and J. D. Reppy, Phys. Rev. Lett. 35, 1286 (1975).
[19]J. E. Rutledge, W. L. McMillan, and J. M. Mochel, in *Quantum Fluids and Solids,* edited by S. B. Trickey, E. D. Adams, and J. W. Dufty (Plenum, New York, 1977).
[20]J. E. Berthold, D. J. Bishop, and J. D. Reppy, Phys. Rev. Lett. 39, 348 (1977).
[21]J. E. Berthold, R. W. Giannetta, E. N. Smith, and J. D. Reppy, Phys. Rev. Lett. 37, 1138 (1976).
[22]J. M. Parpia, D. J. Sandiford, J. E. Berthold, and J. D. Reppy, Phys. Rev. Lett. 40, 565 (1978).
[23]D. J. Bishop and J. D. Reppy, Phys. Rev. Lett. 40, 1727 (1978).
[24]S. Gregory and C. C. Lim, Phys. Rev. A 9, 2252 (1974).
[25]K. R. Atkins, H. Seki, and E. U. Condon, Phys. Rev. 102, 582 (1956).
[26]K. A. Shapiro and I. Rudnick, Phys. Rev. 137, A1383 (1965).
[27]D. F. Brewer, A. J. Symonds, and A. L. Thomson, Phys. Rev. Lett. 15, 182 (1965).
[28]D. F. Brewer, in *The Physics of Liquid and Solid Helium,* edited by K. H. Brennemann and J. B. Ketterson (Wiley, New York, 1978), Part 2.
[29]D. J. Bishop, J. M. Parpia, and J. D. Reppy, in *Low Temperature Physics-LT14,* edited by M. Krusius and M. Vuorio (Elsevier, New York, 1975), Vol. I, p. 380.
[30]M. H. W. Chan, A. W. Yanof, F. D. M. Pobell, and J. D. Reppy, in *Low Temperature Physics-LT13,* edited by K. D. Timmerhaus, W. J. O'Sullivan, and E. F. Hammel (Plenum, New York, 1972), Vol. I, p. 229.
[31]E. N. Smith, D. J. Bishop, J. E. Berthold, and J. D. Reppy, J. Phys. (Paris) 39, C6-342 (1978).
[32]D. J. Bishop, Ph.D. thesis (Cornell University, 1978) (unpublished).
[33]A. Tyler and M. Bagley, J. Low Temp. Phys. 26, 573 (1977).
[34]A. W. Yanof and J. D. Reppy, Phys. Rev. Lett. 33, 631 (1974).
[35]K. R. Atkins, Phys. Rev. 113, 962 (1959).
[36]C. W. F. Everitt, K. R. Atkins, and A. Denenstein, Phys. Rev. 136, A1494 (1964).
[37]A. W. Yanof, Ph.D. thesis (Cornell University, 1975) (unpublished).
[38]J. H. Scholtz, E. O. McClean, and I. Rudnick, Phys. Rev. Lett. 32, 147 (1974).
[39]M. Chester and L. C. Yang, Phys. Rev. Lett. 31, 1377 (1973).
[40]J. G. Dash and J. A. Herb, Phys. Rev. A 7, 1427 (1973); M. W. Cole, J. G. Dash, and J. A. Herb, Phys. Rev. B 11, 163 (1975); J. G. Dash, *ibid.* 15, 3136 (1977).
[41]L. Landau, J. Phys. (Moscow) 5, 71 (1941).
[42]C. G. Kuper, Physica 24, 1009 (1958).
[43]T. C. Padmore, Phys. Rev. Lett. 32, 826 (1974).
[44]T. C. Padmore, Phys. Rev. Lett. 28, 1512 (1972).
[45]T. C. Padmore and J. D. Reppy, Phys. Rev. Lett. 33, 1410 (1974).
[46]H. Haug, J. Low Temp. Phys. 12, 479 (1973).
[47]W. Götze and M. Lücke, J. Low Temp. Phys. 25, 671 (1976).
[48]T. E. Washburn, J. E. Rutledge, and J. M. Mochel, Phys. Rev. Lett. 34, 183 (1975).
[49]M. Chester and L. Eytel, Phys. Rev. B 13, 1069 (1976).
[50]M. W. Cole and W. F. Saam, Phys. Rev. Lett. 32, 985 (1974).
[51]T. P. Chen, M. J. DiPirro, A. A. Gaeta, and F. M. Gasparini, J. Low Temp. Phys. 26, 927 (1977).
[52]W. Thomlinson, J. A. Tarvin, and L. Passell, Phys. Rev. Lett. 44, 266 (1980).
[53]C. J. N. van den Meijdenberg, K. W. Taconis, and R. de Bruyn Ouboter, Physica 27, 197 (1961).
[54]E. L. Andronikashvili, Zh. Eksp. Teor. Fiz. 16, 780 (1946).
[55]M. Chester and L. C. Yang, Phys. Rev. Lett. 31, 1377 (1973).
[56]R. W. Guernsey, Jr., R. J. McCoy, M. Steinback, and J. K. Lyden, Phys. Lett. 58A, 26 (1976).
[57]P. C. Main, C. W. Kieweit, W. T. Band, J. R. Hook, D. J. Sandiford, and H. E. Hall, J. Phys. C 9, L397 (1976).
[58]J. D. Reppy, in *Physics at Ultralow Temperatures,* edited by E. Sugawara (Physical Society of Japan, Tokyo, 1978), p. 89.
[59]J. B. Mehl and W. Zimmerman, Jr., Phys. Rev. 167, 214 (1968).
[60]D. F. Brewer, A. J. Dahm, J. Hutchins, W. S. Truscott, and D. N. Williams, J. Phys. (Paris) 39, C6-351 (1978).
[61]G. Ahlers, in *The Physics of Liquid and Solid Helium,* edited by K. H. Benneman and J. B. Ketterson (Wiley, New York, 1976), Part 1.
[62]K. R. Atkins and I. Rudnick, in *Progress in Low Temperature Physics,* edited by C. J. Gorter (North-Holland, Amsterdam, 1970), Vol. VI, p. 37.
[63]B. Ratnam and J. Mochel, J. Low Temp. Phys. 3, 239 (1970).